REMARKS

Claims 84, 85, 91, 92, 94 and 95 are pending in the Application

Claims 84, 85, 91, 92, 94 and 95 are rejected.

I. REJECTION UNDER 35 U.S.C. § 112, ¶ 1

Examiner has rejected Claims 94 and 95 under 35 U.S.C. § 112, ¶ 1, as failing to comply with the written description requirement. Final Office Action at 2.

The Examiner contends that the instant specification teaches that "[t]he ends of the single-wall carbon nanotubes are open, or closed. However, a combination of open and closed nanotubes is not taught in the specification." *Id.*

Applicant respectfully traverses the rejection.

Support for the combination of open and closed nanotubes is taught multiple times in the Application, for example, as cited below.

> Fullerene tubes may be closed at one or both ends with end caps or open at one or both ends.

Application at 25, *ll.* 16-17.

> The nanotube can have a fullerene cap (e.g., hemispheric) at one end of the cylinder and a similar fullerene cap at the other end. One or both ends can also be open.

Id. at 33, *ll.* 24-26.

> The tubular carbon molecules (including the multiwall forms) produced as described above can be chemically derivatized at their ends (which may be made either open or closed with a hemi-fullerene dome).

Id., at 34, *ll.* 11-13.

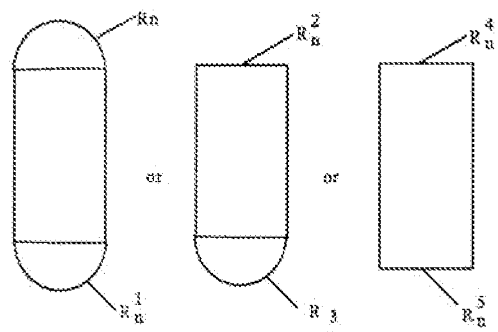
> Alternatively, the fullerene caps of the single-walled nanotubes may be removed at one or both ends of the tubes by short exposure to oxidizing conditions (e.g., with nitric acid or O_2/CO_2) sufficient to open the tubes but not etch them back too far, and the resulting open tube ends maybe

> derivatized using known reaction schemes for the reactive sites at the graphene sheet edge.

Id., at 34, *ll.* 17-22.

In addition, the combinations of end-derivatized single-wall carbon nanotubes are diagrammed, as shown below:

In general, the structure of such molecules can be shown as follows:



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Id., at 34, *ll.* 23-25.

In these three molecules diagrammed in the Application and reproduced above, the vertical rectangular boxes are each a "substantially defect-free cylindrical graphene sheet (which optionally can be doped with non-carbon atoms) having from about 10^2 to about 10^6 carbon atoms, and having a length of from about 5 to about 1000 nm, preferably about 5 to about 500 nm" *i.e.*, the tube section of a single-wall carbon nanotube. *Id.*, at 35, *ll.* 6-10. Moreover, in these three molecules reproduced above, the semi-circles are each "a fullerene cap that fits perfectly on the cylindrical graphene sheet, has at least six pentagons and the remainder hexagons and typically has at least about 30 carbon atoms.," *i.e.*, an end cap of a single-wall carbon nanotube. *Id.*, at 35, *ll.* 12-15. Hence, the second molecule (II) reproduced above is a single-wall carbon nanotube with one end opened and the other end closed.

In light of the foregoing, Applicant respectfully asserts that the claimed subject matter was described in the specification in such a way as to reasonably convey to one skilled in the art

and requests the Examiner withdraw his rejection of Claims 94 and 95 under 35 U.S.C. § 112, ¶ 1.

II. <u>REJECTION UNDER 35 U.S.C. § 103(a) OVER *HIURA* IN VIEW OF *IIJIMA*</u>

The Examiner has maintained the rejection of Claims 84-85, 91-92 and 94-95 under 35 U.S.C. § 103(a) as being unpatentable over *Hiura et al.*, United States Patent No. 5,698,175 ("*Hiura*") in view of Iijima, et al., "Single-shell carbon nanotubes of 1-nm diameter," *Nature*, Vol. 363, June 17, 1993, pp 603-605 ("*Iijima*"). Final Office Action, at 3.

Applicant respectfully traverses the rejection for the same reasons articulated in its Amendment Under 37 C.F.R. § 1.111, filed June 26, 2006 ("the 1.111 Amendment"), at 6-10.

In responding to the Applicant's argument, the Examiner has apparently misunderstood Applicant's position, which has led the Examiner to be un-persuaded. Hence, Applicant is compelled to explain its position with further clarity and then address Examiner's response thereto.

In the 1.111 Amendment, Applicant explained that a person of ordinary skill in the art would have understood that, under the teachings of *Hiura*, the multi-wall carbon nanotubes were damaged by the disclosed process and resulted in the breaking of carbon-carbon bonds generally at regions of defects on the walls and ends of the multi-wall carbon nanotubes. 1.111 Amendment, at 8. Such a person would have further recognized that because of the multi-wall structure of the carbon nanotubes being tested in *Hiura*, those carbon nanotubes would have withstood the attack to their wall layers and would still have remained intact. *Id.* However, and important to the present analysis, a person or ordinary skill in the art would have expected that single-wall carbon nanotubes would have been destroyed by the bond-splitting that was occurring in the *Hiura* process. *Id.* Thus, a person of ordinary skill in the art would have believed that combining *Hiura* with *Iijima* (*i.e.*, performing the *Hiura* process of single-wall carbon nanotubes) would have failed to achieve the desired results. *Id.*, at 8-9. Rather, the results of such a process would reasonably have been believed to be that the *Hiura* process

would destroy the single-wall carbon nanotubes. *Id.*, at 9. In short, the results of the claimed process for the Application yielded a result that a person of ordinary skill in the art would not have expected. *Id.* As real world evidence that the result of the present Application was unexpected, Applicant provided the Examiner with *Dujardin*. *Id.* *Dujardin* showed that, even well after the publication of *Hiura* and *Iijima*, "[i]t was believed that purification of single-shell nanotubes based on preferential oxidation and/or separation using surfactants was much more difficult than that of [multi-shell] nanotubes."

The Examiner asserted two reasons why Applicant's argument was un-persuasive.

First, the Examiner ignored the statement made in *Dujardin* because, according to Examiner, this statement applied to only gas-phase oxidation of multi-shell nanotubes. Final Office Action, at 5. This is not so. As an initial matter, one of the authors of *Dujardin* is Prof. T. W. Ebbesen. *Dujardin*, at 611. Dr. Ebbesen is also, in fact, a named co-inventor of *Hiura*. *Hiura*, at cover page. Hence, there can be no dispute that the authors of *Dujardin* were fully cognizant of the processes disclosed in *Hiura*, when providing this above-quoted statement. However, any question that this statement applied also to the processes disclosed in *Hiura* is completely answered by looking at the endnotes of *Dujardin*.

In particular, at the end of the above-quoted statement in *Dujardin*, the authors referenced four papers, including end note [6], which is H. Hiura, T.W. Ebbesen, K. Tanigaki, *Adv. Mater.*, **1995**, 7, 275. *See Dujardin*, at 611 & 613. Attached hereto at **Exhibit B** is a copy of Hiura *et al.*, "Opening and Purification of Carbon Nanotubes in High Yields," *Adv. Mater.*, 7, No. 3, 275-76 (1995) ("*1995 Hiura*"), which is the paper cited in endnote [6] in *Dujardin*. *Hiura 1995* makes clear the authors (which included both Dr. Hidefumi Hiura and Dr. T.W. Ebbesen, the named inventors of *Hiura)* were trying "well-known oxidants, such as nitric acid, sulfuric acid, the mixture of both and potassium permanganate" for opening and purifying multi-shelled carbon nanotubes. *1995 Hiura*, at 275. Thus, the statement made in *Dujardin* applied to liquid-phase oxidation (as well as gas-phase oxidation) of multi-shell nanotubes. And, therefore,

Dujardin directly showed that a person of ordinary skill in the art would not have expected at the time of the invention the results shown in the present Application. **For that matter, this evidence clearly shows that even the inventors of *Hiura* would not have expected this result.**

The only other basis the Examiner asserted as to why the argument presented by Application was "unpersuasive" was "because the instant process is also carried out by reacting nanotubes with highly oxidative acids and/or oxidation agents under reflux conditions. Therefore, it appears that the process of *Hiura* in view of *Iijima* and the instantly claimed process would produce similar results." Final Office Action, at 4. In other words, the Examiner is relying on Applicant's own disclosure to argue that the unexpected result was expected. It is clearly error to rely on Applicant's own disclosure in this manner. Otherwise, if this were allowed, the results of the applied for invention could always be used to determine what a person of ordinary skill should have expected and, thus, no invention could ever be "unexpected." In short, to protect against hindsight analysis, the Applicant's own disclosure can never be utilized in this manner to show obviousness.

Accordingly, it is impermissible to rely on the Applicant's disclosure to show the invention was expected and the Examiner cannot ignore the statements made in the art that show the a person of ordinary skill in the art would reasonably have been believed the *Hiura* process would destroy single-wall carbon nanotubes.

Evidence of unexpected results must be considered by Examiner. M.P.E.P. § 2141; *see also In re Sernaker*, 702 F.2d 989, 996, 217 U.S.P.Q. 1, 7 (Fed. Cir. 1983). Moreover, such evidence "serve[s] as insurance against the insidious attraction of the siren hindsight" when evaluating the prior art. *W.L. Gore & Assoc. v. Garlock, Inc.*, 721 F.2d 1540, 1553, 200 U.S.P.Q. 303, 313 (Fed. Cir. 1983).

In light of the foregoing, Applicant respectfully requests that the Examiner withdraw the rejection of Claims 84-85, 91-92 and 94-95 under 35 U.S.C. §103(a) as being unpatentable over *Hiura* in view of *Iijima*.

III. CONCLUSION

As a result of the foregoing, it is asserted by Applicant that the Claims in the Application are now in a condition for allowance, and respectfully requests allowance of such Claims.

Applicant respectfully requests that the Examiner call Applicant's attorney at the below listed number if the Examiner believes that such a discussion would be helpful in resolving any remaining problems.

Respectfully submitted,

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EXHIBIT B

Communications

Opening and Purification of Carbon Nanotubes in High Yields**

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Carbon nanotubes can be thought of as cylindrical graphitic micro-crystals with nanometer diameters which are expected to have unique properties.[1] Currently multi-shell nanotubes can be made in gram-orders using the carbon arc discharge synthesis method.[2,3] However, the crude sample contains not only nanotubes but also nanoparticles with a weight ratio of about 2:1 in the best cases. Therefore the nanotubes must be further purified in order to investigate precisely their properties.

Recently we reported the purification of nanotubes by oxidation in air at high temperature (around 750 °C).[4] In this process the nanoparticles are consumed more rapidly than the nanotubes. However the yields are very small (< 1 %) probably due to the local inhomogeneities in the gas phase oxidation process. To overcome this problem, we have investigated the liquid-phase oxidation of the crude nanotube samples and found that under suitable conditions high yields of opened purified nanotubes can be obtained.

In this paper we describe this method which gives yields of the order of 40 % depending on the composition of the crude sample. This method is very useful to obtain gram-quantities of purified nanotubes at one time under well-controlled conditions.

By analogy with the gas-phase oxidation referred to earlier,[4] we tried several well-known strong oxidants, such as nitric acid, sulfuric acid, the mixture of both and potassium permanganate. Transmission electron microscopy (TEM) observation revealed that the oxidation was much more homogeneous in solution where the nanotube sample could be well dispersed through sonication and the chemical attack could be uniform. The oxidation by sulfuric and nitric acids is extremely slow and weak. A mixture of the two gives slightly better results.

For purification purposes, by far the best oxidant is potassium permanganate in acidic solution. Therefore, for the latter case, the methodology will be described in detail next. 1.00 g of the core fibrous material is taken from the deposit synthesized by the carbon arc discharge method,[2,3] ground and dispersed in 200 ml of 1 N sulfuric acid. This solution is placed in a two necked flask fitted with a reflux condenser. Separately, 19.8 g (1.5 molar excess in terms of atomic carbon content) of potassium permanganate is dissolved in 200 ml of 1N sulfuric acid and placed in a funnel fitted to the side neck of the flask. The flask is heated to 150 °C in an oil bath with vigorous stirring (magnetic stirrer). The potassium permanganate solution is then added drop by drop through the side funnel. The solution is then refluxed for an additional 5 hours. The mixture is then cooled and filtered through a G4 glass filter (pore size 10–16 mm). The filtrate is washed with pure water and then with concentrated hydrochloric acid to remove the manganese(IV) oxide (reduced from the original potassium permanganate). When all the manganese oxide has been dissolved away, the filtrate is again washed with pure water. The residual filtrate containing the purified nanotubes is collected and dried under vacuum for several hours.

The yield of the purified nanotubes is about 40 % in terms of the total weight of the starting raw material. Figure 1 shows the TEM pictures of the sample before and after this

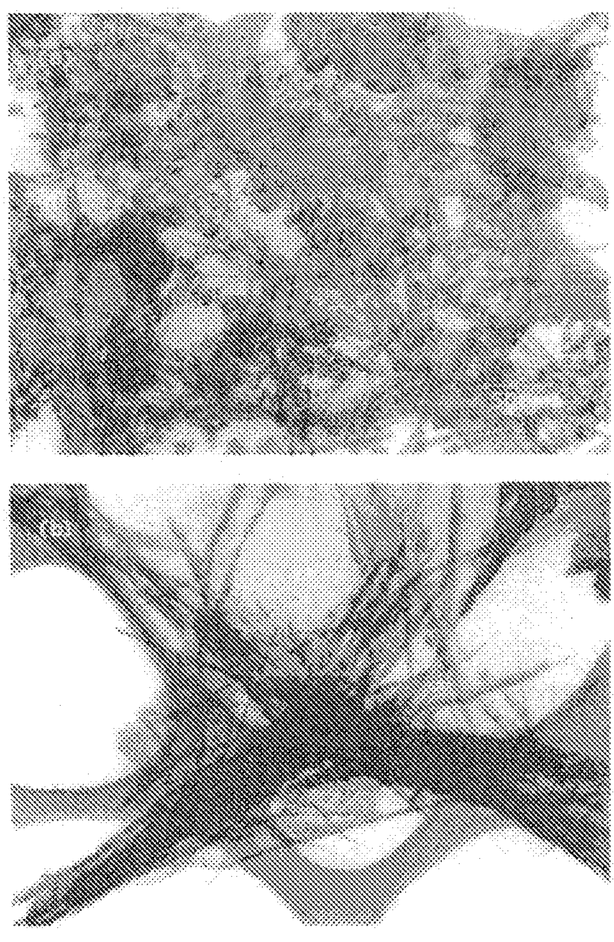

Fig. 1. Carbon nanotubes before (a) and after (b) liquid phase purification.

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[**] We thank M. M. J. Tracey and M. E. Bisher for the micro X-ray analysis of the sample. Since submission of this manuscript, a method for opening nanotubes by nitric acid has been reported (Tsang et al., *Nature* **1994**, *372*, 159) and also confirming that nanotubes are wet by low surface tension liquids such as solvents as predicted in [7].

process. As can bee seen the nanoparticles have been removed. Sometimes some new impurities appear in the sample due to the numerous steps in the process. Micro X-ray analysis indicates one impurity is composed of silicon. This is most likely due to the etching of the glass flask by the boiling sulfuric acid and potassium permanganate mixture. This should be easily removed by treating the sample with HF, or using a different container. The maximum yield cannot be higher than the amount of nanotubes in the original crude sample. Therefore the necessary excess of potassium permanganate with respect to the starting material must be calibrated to the weight ratio of nanoparticles to nanotubes in the sample.

In our case, TEM observations revealed that when the molar ratio is around 0.2, the nanotubes begin to opened and the nanoparticles become round due to reaction at the edges. When the molar ratio is about 1.5, the nanotubes are purified. However if the excess is further increased (e.g. 2.2), the nanotubes become shortened due to the excessive oxidation. The shortening is due to the fact that oxidation starts at the tips of the nanotubes as has been shown elsewhere.[5,6] This is mostly due to strain at the tip which enhance its reaction rate.[1]

The purification process therefore also opens the nanotubes on a large scale. This will be useful for doing nanoscale experiments in the hollow cavities of the nanotubes since we have shown elsewhere that they are wetiable by low surface tension liquids.[7] The oxidation process changes the chemical make-up of the reactive edge of the tips and perhaps even the outer (and the inner) layer of the nanotube. This in turn might affect their properties. To check for any significant chemical modification, the purified nanotubes were analyzed by XPS (ESCA). The detailed results of the XPS analysis will be described elsewhere.[8]

Figure 2 shows the peak-separated difference XPS spectrum of the purified nanotubes around 285 eV. The main

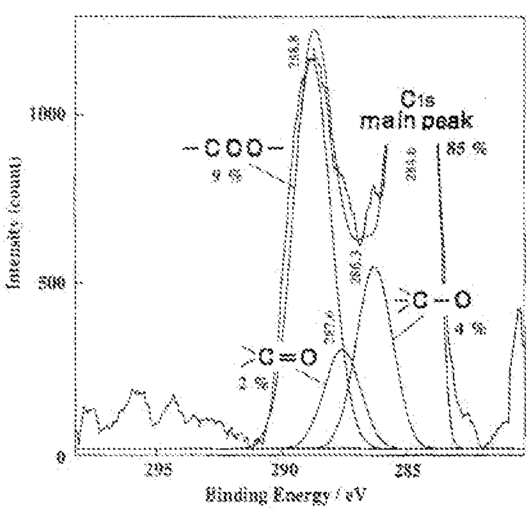

Fig. 2. The peak-separated XPS spectrum of the purified nanotubes around 285 eV. The weight percent of each of the chemical groups present are indicated.

peak (cut off) at 284.6 eV is unambiguously assigned to the C1s of graphitic carbon. The shoulder of the main peak is composed of three peaks, which are assigned to the C1s of hydroxyl carbon (286.3 eV), that of carbonyl carbon (287.6 eV) and that of carboxyl carbon (288.8 eV). The analysis of the peak areas indicates that ca. 15 percent of the carbon constituting the nanotubes are bound to those groups as indicated in Figure 2. Clearly, not only the tips but also the surface of the outer layer is covered with these species. The presence of these hydrophilic group is noticeable in that the purified nanotubes disperse much more easily in water than the original sample.

In conclusion, we have presented a simple method to obtain opened and purified nanotubes in high yields which should allow for their accurate physical characterization. With the resultant higher dispersibility, this method also opens the way for further chemical decoration and modification of carbon nanotubes and their eventual use in biomimetic systems.

Received: October 8, 1994
Final version: December 13, 1994

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[8] H. Hiura, *Mol. Cryst. Liq. Cryst.*, in press.

Photoconductivity in the Columnar Phases of a Glassy Discotic Twin**

By *Dieter Adam, Peter Schuhmacher, Jürgen Simmerer, Lukas Häußling, Wolfgang Paulus, Karl Siemensmeyer, Karl-Heinz Etzbach, Helmut Ringsdorf,* and *Dietrich Haarer**

The electronic charge-carrier mobilities of organic systems are mainly determined by the supramolecular arrangement and packing of the molecular units involved. The overlap of molecular wavefunctions is often more important than the

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[**] We thank P. A. Heiney and N. C. Maliszewskyj for the X-ray measurements, which were carried out in the Materials Research Laboratory at the University of Pennsylvania, supported by NSF-DMR-92-20668. This work was supported by the BMFT (03M4048A0) and by the Fond der Chemischen Industrie.